

## PATENT ABSTRACTS OF JAPAN

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(54) MOISTURE CURABLE TYPE HOT MELT ADHESIVE AND METHOD FOR PRODUCING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a moisture curable type hot melt adhesive capable of being applied on an adherend at  $\leq 100^{\circ}$  C low temperature, and developing a sufficient structural strength after its moisture cure.

SOLUTION: This moisture curable type hot melt adhesive contains a composition obtained by dissolving or dispersing 100-500 pts.mass adhesion-imparting resin dissolvable or dispersible in an alkylalkoxysilyl group-containing polyoxyalkylene at  $100-200^{\circ}$  C based on 100 pts.mass liquid state alkylalkoxysilyl group-containing polyoxypropylene, as a main component.

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CLAIMS

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[Claim(s)]

[Claim 1] Per liquefied alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section, moisture hardening form hot melt adhesive which uses as a principal component the constituent which is dissolved or stabilized in alkyl dialkoxy silyl radical content polyoxyalkylene, and comes to blend the tackifier 100 which can be distributed - the 500 mass sections.

[Claim 2] The manufacture approach of the moisture hardening form hot melt adhesive characterized by being dissolved or stabilized per liquefied alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section and in alkyl dialkoxy silyl radical content polyoxyalkylene, and dissolving or distributing the tackifier 100 which can be distributed - the 500 mass sections, removing moisture at 100-200 degrees C.

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## DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the hot melt adhesive and its manufacture approach of a high adhesive property and the moisture hardenability which has thermal resistance after hardening also to the adherend of a polyolefine system.

[0002]

[Description of the Prior Art] Moisture hardening form hot melt adhesive is widely used as adhesives excellent in the thermal resistance which does not use an organic solvent for fields, such as automobile relation, building materials, and woodwork furniture. Moreover, the reaction hot melt adhesive constituent which comes to blend the tackifier which does not react to the both ends obtained by the reaction of polyolefine mold diol and diisocyanate with the urethane prepolymer which has an isocyanate radical, and this urethane prepolymer, and has compatibility is conventionally well-known at JP.3-111475.A.

[0003] However, this moisture hardening form hot melt adhesive has the fault which does not discover adhesive strength immediately after coating lamination, or stops at low adhesive strength relatively. Moreover, the present condition is that there are still no moisture hardening form adhesives in which the adhesive property which was excellent also even in after hardening to the polyolefine system ingredient is shown.

[0004]

[Problem(s) to be Solved by the Invention] This invention shows sufficient initial adhesive strength to have a tacking function immediately after coating lamination, and it aims at offer of the moisture hardening form hot melt adhesive which discovers a good adhesive property also into polyolefine system ingredients, such as polypropylene, not only immediately after coating lamination but after hardening, and its manufacture approach while it discovers sufficient bond strength as structural adhesive, when a hardening reaction fully advances in the passage of time.

[0005]

[Means for Solving the Problem] This invention is what attains this purpose. Per liquefied alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section. The moisture hardening form hot melt adhesive which uses as a principal component the constituent which is dissolved or stabilized in alkyl dialkoxy silyl radical content polyoxyalkylene, and comes to blend the tackifier 100 which can be distributed - the 500 mass sections. And per liquefied alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section. It is the manufacture approach of the moisture hardening form hot melt adhesive characterized by dissolving or distributing, being dissolved or stabilized in alkyl dialkoxy silyl radical content polyoxyalkylene, and carrying out, moisture removal of the tackifier 100 which can be distributed - the 500 mass sections at 100-200 degrees C.

[0006]

[Embodiment of the Invention] Hereafter, this invention is explained concretely. The liquefied alkyl dialkoxy silyl radical content polyoxyalkylene used by this invention is liquefied polyoxyalkylene containing an alkyl dialkoxy silyl radical, and can mention the polyoxysilylene of

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methyl dimethoxy silyl radical content, polyoxypropylene, a polytetramethylene glycol, etc. the polyoxysilylene of ethyldimethoxy silyl radical content, polyoxypropylene, a polytetramethylene glycol, etc.

[0007] As liquefied alkyl dialkoxy silyl radical content polyoxyalkylene, what contains an alkyl dialkoxy silyl radical in the both ends of polyoxyalkylene is desirable, and that whose number average molecular weight is 1,000 to about 30,000 is desirable.

[0008] The tackifier used by this invention is dissolved or stabilized in said alkyl dialkoxy silyl radical content polyoxyalkylene, and is a tackifier which can be distributed. Monochrome or dialcohol of petroleum resin, hydrogenation petroleum resin, and hydrogenation petroleum resin, Monochrome or dialcohol of rosin resin, hydrogenation petroleum resin, and hydrogenation rosin resin. They are a kind of tackifiers, such as rosin ester resin, terpene resin, terpene phenol resin, and cumerone indene resin, or two sorts or more. These are made into a subject. In compatibility or the range distributed micro Styrene isoprene styrene resin, Styrene styrene-butadiene-rubber resin, hydrogenation styrene isoprene styrene resin, hydrogenation styrene styrene-butadiene-rubber resin, polyisoprene rubber, hydrogenation polyisoprene rubber, polyester resin, an epoxy resin, etc. can be used together.

[0009] The moisture hardening form hot melt adhesive of this invention is obtained by blending the tackifier of per (100) alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section - the 500 mass sections.

[0010] The cohesive force of an adhesives layer runs short of the amounts of the tackifier per alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section under in the 100 mass sections, and bond strength sufficient by the ordinary state is not discovered. On the other hand, if the 500 mass sections are exceeded, the brittleness under the low temperature of an adhesives layer will worsen.

[0011] The moisture hardening form hot melt adhesive of this invention can be manufactured by performing the dissolution or distributed mixing, removing the moisture which exists said alkyl dialkoxy silyl radical content polyoxypropylene and said tackifier in a raw material at the temperature of 100-200 degrees C. In addition, although removal of moisture can be performed by heating and stirring at the temperature of 100 degrees C or more, it is desirable to carry out especially under the circulation condition of inert gas, such as nitrogen gas and carbon dioxide gas, or reduced pressure. In the case of the former, the exclusion effectiveness of the oxidation degradation of adhesives can also be done so. Even if it carries out under reduced pressure, circulating inert gas, of course, it does not interfere.

[0012] When mixing at the temperature of less than 100 degrees C, the dissolution or distributed mixing of alkyl dialkoxy silyl radical content polyoxypropylene, a tackifier, etc. not only takes long duration, but removal of moisture may become inadequate under reduced pressure. On the other hand, although the dissolution or distribution becomes easy in mixing at the high temperature exceeding 200 degrees C, the pyrolysis of dissolution mixture becomes remarkable and there is also concern which spoils a fluidity by gelation.

[0013] In the moisture hardening form hot melt adhesive of this invention, fillers, such as a silica, a kaolin, talc, and a calcium carbonate, a phthalic-acid system plasticizer, an adipic-acid system plasticizer, a phosphoric acid system plasticizer, liquid rubber, process oil, extender oil, etc. can be added for the improvement of the flowability at the time of adhesive ability or melting. [0014] moreover, to the moisture hardening form hot melt adhesive of this invention in order to promote hardening of alkyl dialkoxy silyl radical content polyoxyalkylene Dibutyltin dilaurate, Dibutyltin malate, Dibutyltin acetate, Organic tin compounds, such as Dibutyltin acetate, acetylacetonate, tetrabutyl titanate, Organic titanium compounds, such as tetra-propyl titanate, aluminum thoria cetyl acetate, Organometallic compounds, such as organoaluminum compounds, such as aluminum-triethyl acetate and JISOPUROPOKISHI aluminum ethyl acetate, can be added as a curing catalyst. It is desirable to use 0.1 - 5 mass section to the alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section.

[0015] Furthermore, it is desirable to be able to add silane coupling agents, such as vinyltrimethoxysilane, N-(beta-aminoethyl)-gamma-aminopropyl dimethoxysilane, gamma-aminopropyl trimethoxysilane, and gamma-aminopropyl triethoxysilane, to the moisture hardening

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form hot melt adhesive of this invention, and to double and use the 0.1-10 mass section for it to the alkyl dialkoxy silyl radical content polyoxyalkylene 100 mass section, in order to improve an adhesive property and storage stability.

[0016]

[Example] Hereafter, an example explains this invention still more concretely. In addition, the engine performance of the obtained moisture hardening form hot melt adhesive was measured by the following approach.

[0017] 1. the melt viscosity of melt viscosity profit \*\*\*\* moisture hardening form hot melt adhesive was measured at 80 degrees C using the Brookfield viscometer.

[0018] 2. Fill Up 300 Cc Cartridge made from Aluminum with Moisture Hardening Form Hot Melt Adhesive, and Heat at 100 Degrees C. Adhesion between Ordinary State Reinforcement and Strength-in-High-Temperature (1). Griddle and Canvas - \*\*\*\*\* - Having Had - It applies to the thickness of about 100 micrometers by 25mm width of face on a 2mm griddle using cartridge gun ETR-100 made from SANTSURU / D. Moreover No. 9 canvas was made to rival and it pressed for 10 seconds by the pressure of 53kPa(s), it was recuperated at the room temperature for 24 hours, and the specimen was created, and using this, the peel strength (unit: N/25mm) for 5m/in tension rate was measured, and it expressed as the room temperature as ordinary state reinforcement. In addition, the value similarly measured in the 80-degree C ambient atmosphere was displayed as strength in high temperature.

(2) The specimen was created and measured like the above except having replaced with .PP (polypropylene) plate and the bond strength griddle between canvas (2mm in thickness), and having used PP plate (2mm in thickness).

[0019] 3. After aging the specimen (a griddle / adhesives / canvas, or PP plate / adhesives / canvas) of the cold-resistant above at minus 30 degree C for 24 hours, the hard ball with a diameter of 15mm was dropped from height of 1.5m on the griddle or PP plate surface, and the damage of each adhesives layer was observed.

O : there is no damage to an adhesives layer and the stable adhesion is maintained.

xx: Adhesion is maintained although a crack is accepted in an adhesives layer.

x: An adhesives layer is destroyed and exfoliation is accepted between adherends.

[0020] Having taken to the separable flask in the amount which shows the SAT350 (Kaneka Co., Ltd. make, design number average molecular weight 9,000) 100 mass section as examples 1-3 and the example 1 of reference, and 2 alkyl dialkoxy silyl radical content polyoxyalkylene (POA), and shows KE-601 (hydrogenation rosin diol by the Arakawa chemical-industry company) in Table 1 as a tackifier, having circulated nitrogen gas at 150 degrees C, and removing moisture, it heated and agitated and the constituent was prepared for 1 hour. Then, after cooling to 80 degrees C, circulating nitrogen gas, moisture hardening form hot melt adhesive with the transparent melting viscosity which blends the Dibutyltin acetate 2 mass section as a curing catalyst, blends the vinyltrimethoxysilane 2 mass section and the gamma-aminopropyl ethoxy silane 3 mass section (5 in all mass section) as a silane coupling agent, and is shown in Table 1 was prepared. The result shown in Table 2 using this moisture hardening form hot melt adhesive of having excelled was obtained.

[0021]

[Table 1]

	配合比 (質量部)				溶融粘度 mPa.s
	POA	粘着剤	硬化剤	付着剤	
実施例 1	100	150	2	5	1,950
実施例 2	100	250	2	5	4,370
実施例 3	100	350	2	5	8,120
比較例 1	100	50	2	5	750
比較例 2	100	650	2	5	31,980

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[0022]

[Table 2]

試料	鉄板とキャンバス			PP板とキャンバス		
	常温粘度	耐熱性	耐水性	常温粘度	耐熱性	耐水性
実施例 1	31.2	8.4	○	11.9	6.2	○
実施例 2	47.2	24.7	○	34.8	12.2	○
実施例 3	58.0	29.8	○	44.3	17.5	△
比較例 1	5.6	2.8	△	4.5	2.6	×
比較例 2	68.8	21.3	×	30.4	16.2	×

[0023] Except having replaced with example 4KE-601 and having used YS-2130 (Yasuhara Chemical terpene phenol resin), like the example 2, it was transparent and the result which prepares the moisture hardening form hot melt adhesive of melt viscosity 19,400mPa.s, and shows it in Table 3 using the obtained moisture hardening form hot melt adhesive of having excelled was obtained.

[0024] Except having replaced with example 5KE-601 and having used KE-100 (hydrogenation rosin resin by the Arakawa chemical-industry company), it was transparent like the example 3 and the result which prepares the moisture hardening form hot melt adhesive of melt viscosity 9,300mPa.s, and shows it in Table 3 using the obtained moisture hardening form hot melt adhesive of having excelled was obtained.

[0025] Except having replaced with example 6KE-601 and having used the neo polymer 120 (petroleum resin by the Nippon Oil chemistry company), it was transparent like the example 2 and the result which prepares the moisture hardening form hot melt adhesive of melt viscosity 7,320mPa.s, and shows it in Table 3 using the obtained moisture hardening form hot melt adhesive of having excelled was obtained.

[0026] The result which prepares the moisture hardening form hot melt adhesive of melt viscosity 3,800mPa.s which became cloudy in homogeneity, and shows it in Table 3 like an example 2 using the obtained moisture hardening form hot melt adhesive of having excelled was obtained except having replaced with example 7KE-601 and having used KR-1840 (hydrogenation petroleum resin diol by the Arakawa chemical-industry company).

[0027]

[Table 3]

試料	鉄板とキャンバス			PP板とキャンバス		
	常温粘度	耐熱性	耐水性	常温粘度	耐熱性	耐水性
実施例 4	38.7	22.5	○	24.3	12.8	○
実施例 5	72.4	40.8	○	51.9	28.4	○
実施例 6	57.7	36.6	○	48.2	26.5	○
実施例 7	43.0	28.7	○	31.1	14.4	○
実施例 8	62.9	34.6	○	51.2	30.3	○

[0028] Example 8KE-601 it replaces with the 250 mass sections and is KE-601. The result which prepares the moisture hardening form hot melt adhesive of melt viscosity 5,580mPa.s, and shows it in Table 3 using the obtained moisture hardening form hot melt adhesive like an example 2 of having excelled was obtained except having used the 150 mass sections and the styrene resin 3100 (product made from HAKYUURESU) 100 mass section.

[0029] Except having made the melting temperature of KE-601 to example of comparison 1SAT350 into 250 degrees C, although carried out like the example 1, it thickened remarkably 1 hour after, and stirring became difficult and stopped.

[0030] Although the dissolution of KE-601 to example of comparison 4SAT350 was performed

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like the example 1 at 80 degrees C except having carried out heating stirring for 8 hours, as shown in Table 4, the moisture hardening form hot melt adhesive of melt viscosity 1,850mPa.s was obtained. Although the result of having measured the melt viscosity in 80 degrees C of 4 hours, 8 hours, and 24 hours after about these adhesives was shown in Table 4, since moisture remained, it turned out that melt viscosity is changed sharply and there is no storage stability. In addition, although the result similarly measured about the moisture hardening form hot melt adhesive prepared in each example for reference was shown in Table 4, there was little change of melt viscosity.

[0031]

[Table 4]

	初期	4 時間後	8 時間後	24 時間後
比較例 4	1,850	4,740	8,550	38,600
実施例 1	1,950	1,960	2,000	2,010
実施例 2	4,770	4,340	4,380	4,380
実施例 3	8,120	8,150	8,200	8,220
実施例 4	19,400	19,900	20,200	20,230
実施例 5	9,300	9,340	9,270	9,400
実施例 6	7,220	7,310	7,400	7,390
実施例 7	3,800	3,770	3,830	3,870
実施例 8	5,580	5,550	5,610	5,640

[0032]

[Effect of the Invention] It can apply to adherend easily at the low temperature of 100 degrees C or less, and can paste up, the moisture in air is absorbed, hardening progresses, and the moisture hardening form hot melt adhesive of this invention discovers structure reinforcement. In addition, a good adhesive property is shown also to the polyolefine ingredient like polypropylene.

[Translation done.]